

The manuscript, "Revolutionizing Hailstone Analysis: Exploring Non-Soluble Particles through Innovative Confocal Laser and Scanning Electron Microscopy Techniques," is an account of investigating hailstones by applying multiple analytical techniques and employing a special FORMVAR coating procedure to preserve the spatial distribution of particles captured within hailstone thin sections. While the manuscript makes several interesting points it suffers from some significant shortcomings. The most concerning of these in my opinion is that the manuscript really focuses on findings from what appears to be one thin section of one hailstone. It is very unclear how representative the results are and moreover, if the focus is more to present the methodology, they do not inspire belief that these types of experiments would be easy and straightforward to reproduce in a manner that would lead to statistically significant data.

Thank you for your review of our work. We understand the initial concern about showing results from one hailstone. We intended to include results comparing two hailstones for a follow-on paper, focusing on methodology. Still, we also recognize the need here to show confidence in the reproducibility of this technique. With the remaining laboratory hours available during this funding cycle, we completed an additional analysis to address your points below. Responses to your suggestions for improvement are provided below, including new figures from the analysis of new stones/stone sections.

Several areas for improvement are:

1. The hailstone preparation and sublimation method could benefit from a better descriptive illustration/figure. The utilization of FORMVAR seems to be a legacy technique that it is not common so that readers might have intuition about how it works.

Thank you for your observation. We do not have additional photos taken during the preparation phase, but we acknowledge the need to provide more detail on this technique. To this end, the following description has been added to the [text in lines 171 through 175](#):

“After taking pictures of the hailstone sample after being polished, a layer of FORMVAR solution is applied to the sample's surface using a clean glass rod. This application is done in two ways: 1) by dipping one side of the rod into the FORMVAR solution and spreading a small amount over the surface, or 2) by pouring small amounts of the solution onto the surface and evenly spreading it across the polished hailstone. Once the entire surface is covered with the FORMVAR solution, the sample is ready for sublimation.”

2. Links between particles and nucleating particles are quite tenuous. There are many particles in the analyzed sample and it appears impossible to deconvolute what was there when the ice began to form, versus what was accumulated during transport in the cloud etc.

Thank you for your comment. We acknowledge that distinguishing between particles present during the initial formation of the hailstone and those accumulated during transport is challenging. However, our approach to distinguishing which particles existed within the

hailstone's core or embryo is meant to highlight which particles were present where nucleation occurred and, thus, were likely involved in the initial nucleation process. These particles are distinguished from those in the outer layers more prone to being acquired during transport. A comparison of particles in the embryo versus outer layers will be shown in a follow-up paper and linked to likely sources in the region for separate hail events. Previous work in melting stones had isolated the embryo (e.g., 3 hailstones melted and analyzed in Michaud et al. 2014) to also make inferences on the composition of particles that have the potential to act as nucleation sites owing to their presence in the embryo. By avoiding the melting stage, we also preserve the in situ location of the particles with respect to the embryo and their sizes and shapes, thus advancing our knowledge of the types and characteristics of particles likely leading to ice nucleation in the formation of hailstones.

3. Figure 9 appears to be the most interesting result, but is difficult to interpret and the photographs that are included are extremely small.

Thank you for this observation. After reviewing Figure 9 and the information we aim to present, as well as taking into account your other suggestions to analyze a different cross-section of the same hailstone as well as other hailstones, we have decided to remake the figure and distribute the information across several new figures as follows:

Our new Figure 9 more clearly shows the two cross-section areas chosen to analyze particle size distribution and composition.

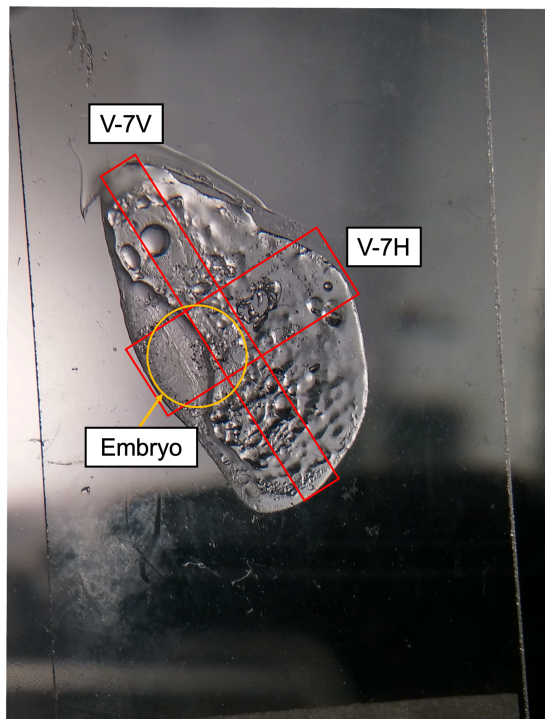


Figure 9: An example analyzed hailstone (V-7) where the areas highlighted by red rectangles indicate where particles were randomly selected to measure particle size distribution using CLSM and elemental composition via SEM-EDS. The orange circle marks the location of the embryo.

A new Figure 10 simplifies the message about the particle size distributions in comparing them among the two cross-sections in Figure 9-A,B (V-7V, V-7H) and two additional stones collected from the same storm (Figure 9-C,D; V-16, V-17). Note that the different axis ranges in this figure represent the different number of particles analyzed and differences in size ranges owing to the different sizes of the hailstones and, thus, cross sections. The results of this new figure are described below in response to your related suggestion to analyze additional stones.

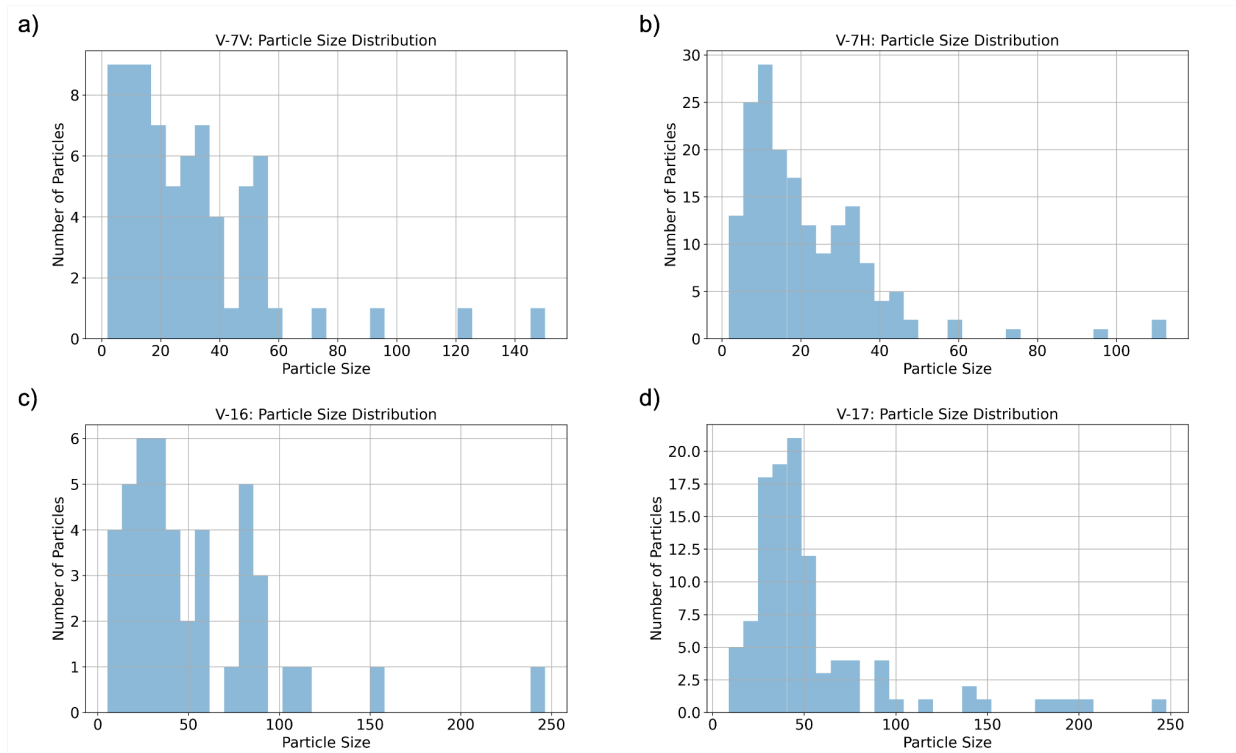


Figure 10: Particle size distributions for hailstone samples collected during the event on 8 February 2018. Panels a) and b) display particle sizes for two different cross-sections from sample V-7, while panels c) and d) show particle sizes for samples V-16 and V-17, respectively.

In our original Figure 9, the details of the composition of particles were obscured by overlapping dots; thus, we have added a new Figure 12 to the manuscript. This figure shows the elemental composition distribution of particles for V-7V (Figure 12-A) and V-7H (Figure 12-) cross-sections (i.e., different cross-sections within the same stone). This figure more clearly demonstrates the dominant elemental compositions within the stone. It shows

that the relative contributions of different compositions are similar when analyzing two different cross-sections of the same sample (i.e., V-7V, V-7V).

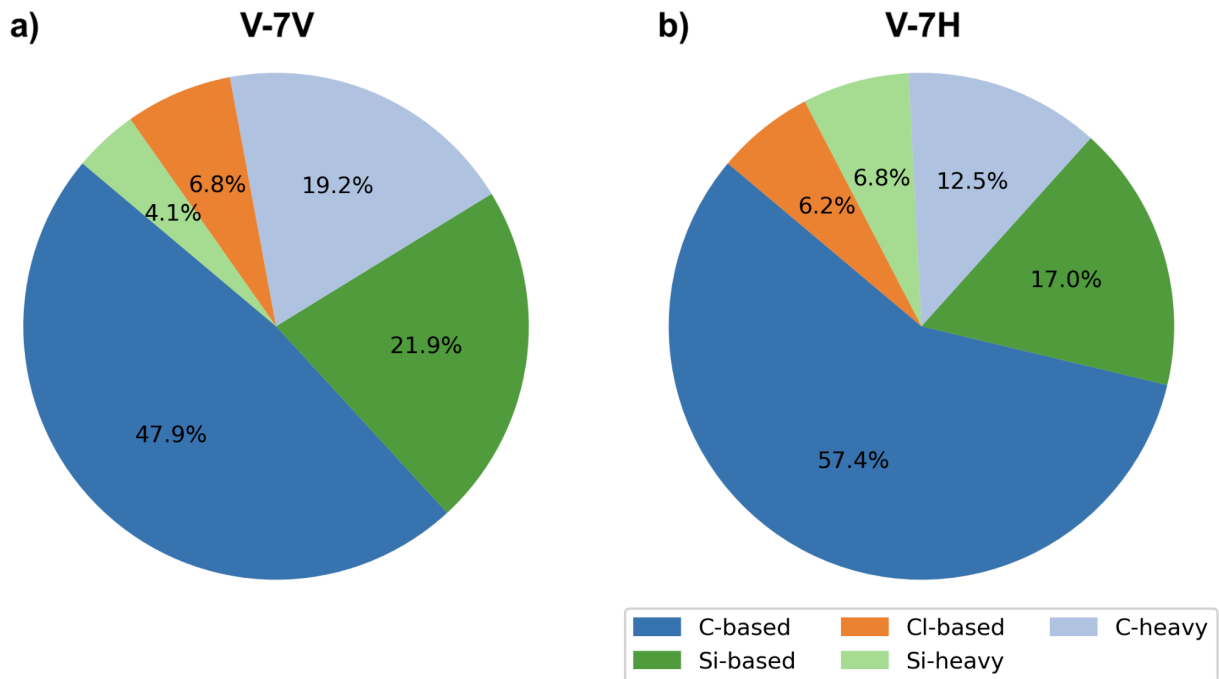


Figure 12: EDS-based elemental composition distribution of particles for a) V-7V and b) V-7H cross-sections, as seen in Figure 9.

Finally, we would like to highlight a benefit of this method in that we can isolate particles within the embryo compared to the outer layers, and in our case, as previously highlighted, being able to describe both the sizes and composition of the particles that may have served in the nucleation process of this stone. Our new Figure 13 isolates just those particles in the embryo regions of the cross sections. Both V-7V (Figure 13-A) and V-7H (Figure 13-B) cross through the embryo (see new Figure 9). Still, different particles were selected within the embryo sample to elucidate better the range of particle characteristics observed within this stone's embryo. Also, because the lead author has gotten more efficient at this process, he was able to analyze more particles in a similar amount of time, thus the difference in particle numbers in the new V-7H analysis. The messages from these figures are described below in response to another of your suggestions.

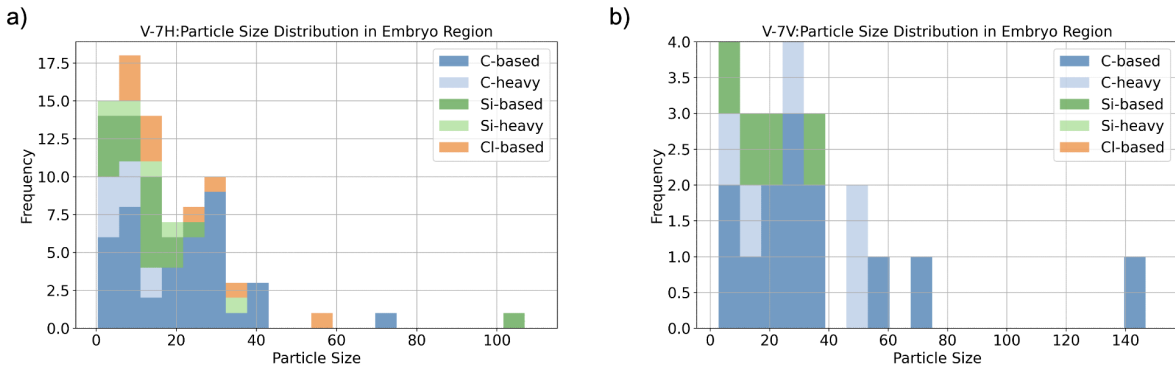


Figure 13: EDS-based elemental composition distribution of particles found in the embryo for sample V-7 for a) V-7V and b) V-7H.

With these new figures, we address your remaining comments below and have enhanced the interpretability of the information presented in the results section of the revised manuscript.

4. Given the lack of duplicates etc. (see comment above) it is very hard to assess the utility of all of the effort that went into this analysis. If one were to take a second thin section of the same stone and repeat the analysis, would we get wildly different results, or similar? What do we learn in either case? What about with another stone from the same storm? Is it even practical to do this work on many stones?

Thank you for this suggestion. Thankfully, we had remaining lab hours budgeted to analyze the particles in a horizontal section through V-7 (V-7H as in new Figure 9). We analyzed particles within the same sectors covering the embryo as in our original map (i.e., V-7V as shown in new Figure 9), including CLSM-based size distributions and EDS-based elemental composition. Due to increased proficiency with this technique, we could analyze more particles in a shorter amount of time. The results of this second cross-section are shown in the revised manuscript as new Figures 9, 10, 12, and 13. Furthermore, we had two other hailstones from this same storm (V-16, V-17) that we could analyze in the size distribution from the CLSM for comparison with the V-7 stone, with results included in the new Figure 10.

Our analysis of this different section in the same hailstone revealed that particles were overall smaller in the V-7H (Figure 10-A) cross-section than in V-7V (Figure 10-B) but still contained a few isolated particles exceeding 100 microns. Additionally, compared to V-16 (Figure 10-C) and V-17 (Figure 10-D), particle sizes in V-7, the smallest hailstone of the three, are relatively smaller (Figure 10). Regarding elemental composition (Figure 12-A,B), we did not find significant differences; carbonaceous particles remained predominant, with silicates being the second most dominant particles. In the embryo region (Figure 13), we discovered that while salts were not identified in the initial analysis of V-7V (Figure 13-A), they were present in the additional horizontal cross-section (Figure 13-B; V7-H), along with

heavier metals. These new figures and their interpretation are included in the Results section of the revised manuscript.

In summary, these figures demonstrate the robustness of our method, showing overall consistent messages but highlighting the value of looking at multiple cross sections in one stone, particularly for identifying a variety of elemental components. Across hail stones from the same storm, there appears to be an increase in particle sizes with increasing size of the hailstone that is an interesting result to explore further, showing the value of this unique method that preserves particles for analysis of both size and composition with respect to the embryo. Although we only show the size and composition within the embryo region here, we have extended this analysis to compare with the outer layers and in comparing results with a hailstone from a different storm under different environmental conditions, and therefore, regional transport sources that are the subject of a soon-to-be submitted paper further highlighting results of this unique method.

5. The CLSM work that lays the foundation for SEM analysis appears to resolve particles down to 1 micron. This is still quite a large size, and many particles will be much smaller than this. Even ice nucleation parameterizations are largely based on particles with sizes greater than 0.5 microns. Many such particles missed here.

We acknowledge that this CLSM analysis will result in missing particles smaller than 1 micron, including those down to 0.05 microns that may serve as INPs. However, the flexibility of our method allows for the analysis to begin with SEM, which provides higher resolution and ensures these smaller particles are not overlooked. Although SEM cannot provide topographical information of the particles, starting with SEM enables the detection of particles smaller than 1 micron. We started with CLSM to get more detail about the individual particles' size/shape/topography. Also, a limitation of analyzing smaller particles with the SEM is that this method requires a glass substrate, which may introduce spectral contamination at smaller sizes. These limitations are included in the revised manuscript between lines 242 through 252.

My overall reaction to the submitted manuscript is that in its current form the work falls short of a new atmospheric measurement technique, or some protocol that could be widely adopted. Rather it is a report on the application of several analytical methods to a single hailstone (or single thin section from a single hailstone) from a unique event. The authors mix cases and do at times refer to the plural "hailstones". If they have more data, I would encourage them to complement the manuscript to find more general conclusions. Without this I do not see the extension to the interests of a general readership. That said, I do complement the authors on the incorporation of citizen science.

Thank you for your comments and the opportunity to address your concerns.

While we primarily discuss a singular hailstone in this manuscript to describe the method proposed for hailstones, we have added more information from another section of the same hailstone and results from two other hailstones from the same storm. Our intention here in this paper is to describe the methodology in detail, its adaptability, limitations, and potential for broader applications, with more results from hailstones in multiple events being the focus of a separate detailed results paper given the length of this detailed methodology.

In response to your observations:

1. We have improved our efficiency with the technique, enabling us to analyze twice as many samples as before in the same amount of time. This increased proficiency demonstrates the potential for scalability and broader application of our methodology.
2. We acknowledge the limitations of the current dataset due to time constraints. However, the lead author is committed to continuing this work post-PhD, further refining the methodology and expanding the dataset to draw more general conclusions for hailstones within and outside Argentina (i.e., in the U.S.).
3. We believe that with practice and incorporating lessons learned as detailed in this methodology-focused manuscript, others can replicate and build upon our methodology. We are open to sharing our experiences and providing guidance to facilitate other researchers' adoption of this technique following the publication of this paper.

We appreciate your recognition of our incorporation of citizen science and will strive to continue engaging with the broader public and scientific community.